



MOVEMENT OF RUTHENIUM IN THE BED OF WHITE OAK LAKE

by

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Currently, several thousand curies per year of ruthenium flow onto the bed of former White Oak Lake from the Oak Ridge National Laboratory's intermediate-level waste pits. As the waste water traverses the lake bed, more than half of the ruthenium is removed from solution. The ruthenium that is not sorbed on the lake-bed soil drains into White Oak Creek, a tributary of the Clinch River.

An investigation was made to determine the quantity and distribution of ruthenium in the soil of the lake bed and to identify and define geo-hydrological factors affecting the movement of ruthenium through the lake bed. As of February 1962, the lake bed contained approximately 1200 curies of ruthenium. The ruthenium is present mainly in two tracts of contamination, covering approximately 10 acres, that coincide roughly with the surface flow of waste over the bed. The highest concentrations of ruthenium occur in the uppermost few inches of the lake bed, and about 70% of the activity is in the top 2 feet of soil.

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The lake bed is underlain by a thin layer of recent lacustrine sediment, several feet of alluvium, and the Conasauga shale formation of Cambrian age. Water-level measurements indicate that the depth to ground water varies from < 1 to 5 feet below the surface. The subsurface migration of ruthenium follows closely the paths indicated by water-table contours. The rate of ground-water movement in the upper 2 feet of soil varies from 1 to 5 feet per day, while movement in the material 2 to 5 feet below the surface ranges from 0.05 to 0.25 feet per day. Thus, the maximum rate of travel of ruthenium in the upper layers of soil is approximately 20 times that of the lower layers.

Only a small fraction of the ruthenium that enters White Oak Creek from the lake bed is transported by ground water through the lake-bed soil into the creek. The ruthenium that is not sorbed moves at such a slow rate through the soil that radioactive decay reduces the concentration of that reaching the creek to insignificant proportions. The amount of surface flow and, consequently, the quantity of ruthenium that reaches the creek from the lake bed varies seasonally. During the dry summer months, drainage from the waste pits recharges the ground water in the lake bed, and, thus, there is little surface flow and, consequently, little ruthenium that flows into White Oak Creek. However, in the wet winter season surface runoff from the lake bed is high, and, therefore, larger amounts of ruthenium enter White Oak Creek.

Introduction

White Oak Lake, shown on Fig. 1, formerly served as a final settling basin for low-level radioactive wastes discharged from ORNL. White Oak Dam, a highway fill located 0.6 mile upstream from the confluence of White Oak Creek and the Clinch River, was closed in October 1943. At full pool the lake extended about 1 mile upstream and covered an area of about 44 acres. The temporary holdup, provided by the impoundment, afforded some dilution and a period for the decay of short-lived radionuclides before release to the Clinch River. It also allowed the deposition and accumulation of contaminated sediments. In 1955 the lake was drained, leaving behind approximately 1,000,000 cubic feet of contaminated lacustrine deposits. Currently, several thousand curies per year of ruthenium flow onto the upper part of the now dry lake bed from the nearby intermediate-level chemical waste pits. As the waste water traverses the lake bed, a large part of the ruthenium is removed from solution. The quantity, distribution, and migration of this ruthenium within the lake bed is discussed in this report.

Intermediate-Level Liquid Waste Disposal at ORNL

At ORNL intermediate-level radioactive liquid waste is discharged to the soil by means of a series of seepage pits located in Melton Valley (See Fig. 1.).^{1, 2} Through December 1961, the system had received about 22 million gallons of waste containing approximately 235,000 curies of Ru^{106} , 183,000 curies of Cs^{137} , 42,000 curies of Sr^{90} , the rare earths, and lesser amounts of Ru^{103} , Sr^{89} , Co^{60} , and Sb^{125} . All of the radioactive materials are not retained by the waste-pit system, and, consequently, some activity is transported by ground water through the soil to surface streams east and west of

the pits. For the period of June 1952 to December 1958, less than 500 curies of activity had migrated from the waste pit area; however, due to a significant increase in the amount of fission products released to the pits during 1959, an estimated 13,000 curies of Ru^{106} was transported from the waste pits onto the lake bed for the 3-year period, 1959-1961. The total amount of all other radionuclides leaving the pit area since the pits were opened is estimated to be less than 250 curies, most of which is Co^{60} .

Geology

The bed of former White Oak Lake is underlain by a thin layer of recent lacustrine sediment, 6 to 12 feet of alluvium, and the Conasauga shale formation of Cambrian age. The lacustrine sediment, which was deposited during the 1949-1955 impoundment, is approximately 2 feet thick in the lower part of the lake bed near the dam; however, there is a gradual thinning of the layer upstream and, in general, toward the shore line of the lake. Physical analysis of this material indicates a composition of approximately 5% sand, 60% silt, and 35% clay and, thus, is classified as a silty clay loam.³ The alluvial material consists of a complex mixture of sand, silt, clay, and gravel. Core and auger samples taken along transects at the lower, middle, and upper parts of the lake showed that there are no beds that persist vertically or laterally. However, detailed mechanical analyses of the core and auger samples indicate that the amount of sand and gravel increases slightly with depth, while the silt content decreases. Too, sand and gravel appear to be more prevalent near White Oak Creek which meanders through the area. There is a lack of bedding in the alluvium, since the material was transported into the area by White Oak Creek and by tributary streams that drain

the surrounding highlands. Near the lower end of the lake, where the alluvium is thickest, well-rounded pebbles and cobbles of quartzite and other resistant materials occur intermixed with the sands, silts, and clays. These pebbles and cobbles, which are not representative of the surrounding bedrock, are believed to be remnants of an ancient flood plain of the nearby Clinch River.

The Conasauga shale formation underlying Melton Valley is about 1500 feet thick, and the most common angle of dip is 30° to 40° to the southeast. It consists essentially of a basal red shale that grades upward into gray calcareous shale containing thin-to-medium beds of limestone. The amount of limestone increases toward the southeast and is predominant in the uppermost portion of the formation.⁴ White Oak Lake lies in the upper part of the formation. Here it consists of gray, thin-to-medium bedded, dense-to-crystalline limestone that is interbedded with buff-colored calcareous shale.

Hydrology

All drainage from the bed of former White Oak Lake is into White Oak Creek, which discharges into the Clinch River, 0.6 mile below White Oak Dam. Backwater from Watts Bar Dam, located on the Tennessee River approximately 60 miles downstream, extends into the lower reaches of the creek during the summer months. Thus, in order to prevent the reservoir water from entering White Oak Creek, the gate at White Oak Dam is set a few inches above the full pool level of Watts Bar Reservoir. The 100,000-cubic-foot impoundment created behind White Oak Dam covers about 20% of the area of the previous (1943 to 1955) inundation.

A series of 31 shallow observation wells was completed in the bed of the lake to define the configuration of the water table and determine the direction of ground-water movement. Water-level measurements in these wells, made about once each week for the period of August 28, 1961, to January 5, 1962, indicate that the depth to ground water in the area generally varies from 3 to 5 feet below the land surface during the dry summer months and from < 1 to 1.5 feet during the wet winter months when transpiration and evaporation are lower.

The general direction of ground-water movement is toward White Oak Creek in the lower portion of the lake bed; however, in the upper bed the movement is more complex. Water-table contour maps indicate that the configuration of the water table in the upper part of the area is measurably affected by the flow of surface water onto the lake bed from the two streams that drain the waste pit area (See Fig. 2.). During the dry summer months these streams recharge ground water in the lake bed, and little or no surface water flows directly into the creek. However, during the wet, winter months, when the ground water in the lake bed is close to the land surface, some of the water from these streams flows over the surface of the lake bed into White Oak Creek.

The hydraulic conductivity or permeability of soil in the lake bed was measured by augering a cavity into the lake bed below the water table, allowing the cavity to fill with water, lowering the water level in the hole by pumping, and observing the rate of rise of water level in the cavity. The rate of rise of water level can be converted by a suitable formula to the hydraulic conductivity (k) for the soil.⁵ Results of a series of such tests

showed that the hydraulic conductivity of the soil in the lake bed was relatively uniform.

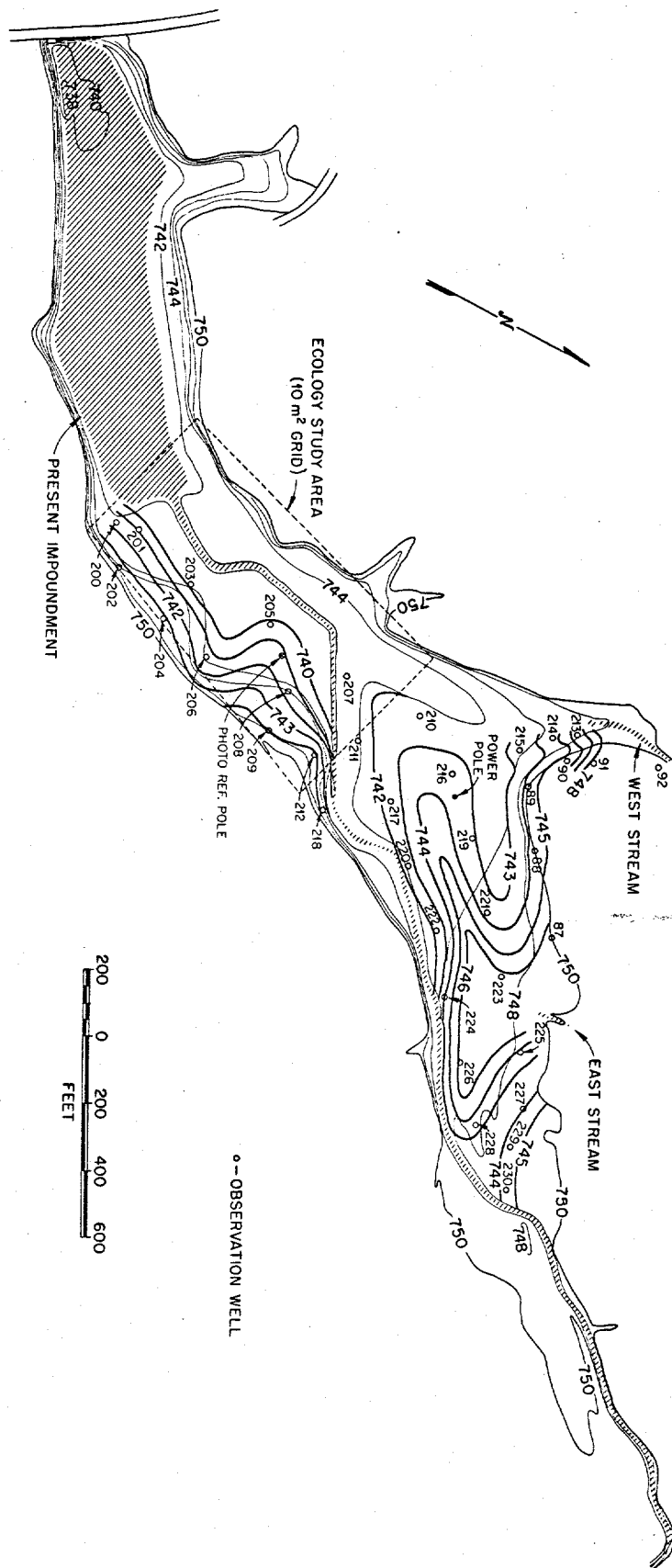


Fig. 2. Water-Table Contours at White Oak Lake Bed, September 22, 1961.

than the underlying material. The average hydraulic conductivity of the upper 2 feet of soil was calculated to be 19 feet per day, whereas the average hydraulic conductivity of the material 2 to 5 feet below the surface was found to be 1 foot per day.

Water-table contour maps were used to determine hydraulic gradients in the lake bed. Figure 2 shows that the hydraulic gradient varies considerably within the area, being lowest in areas where the water table was closest to the land surface. Hydraulic gradients beneath the old lake bed vary from 0.05 to 0.01.

The rate of water movement in the lake bed can be measured by an application of Darcy's law which is

$$V = \frac{ks}{p}$$

where

V = the velocity of flow,

s = the hydraulic gradient,

k = the proportionality factor or the hydraulic conductivity of the porous medium, and

p = the porosity of the material.

By using the measured values given above and by assuming an average porosity of 10%, the average velocity of ground water in the upper and lower layers of lake bed soil were calculated. In the upper 2 feet of soil, average water velocities range from 1 to 5 feet per day, whereas the rate of ground-water movement in the material 2 to 5 feet below the surface varies from 0.05 to 0.25 feet per day. Thus, the average rate of ground-water movement in the upper part of the soil is approximately 10 times that of the lower.

Soil Sampling

General

Due to the nature of the lake bed environment, several obstacles were encountered that hindered soil-sampling operation, as well as other investigations in the area. The entire bed of the lake comprises a terrestrial ecological research site. Therefore, it was essential that sampling operations should not change the environment. In addition, the marshy conditions over much of the site prevented the use of heavy mechanical equipment. Due to the high-radiation field that exists in the area (Radiation measurements made with a portable ionization chamber-type instrument, "cutie pie," show maximum dose rates of 2 rad per hour.), it was necessary to restrict working time.

A 2-inch-diameter thin wall tube, known as the Shelby Tube Sampler, was used to obtain core samples in the lake bed. The instrument was driven into the earth and recovered manually. A total of 54 cores were taken, approximately 50 feet apart, along lines estimated to be at right angles to the surface flow of waste over the lake bed (See Fig. 3.). The contaminated zone encompasses an area of about 10 acres. As previous work indicated that most of the contamination was associated with the upper layer of soil, at each cross section one borehole was sampled to a depth of 5 feet, while the remaining cores were taken to a depth of 2 feet. The samples were sliced and segmented into increments of 0 to 2 inches, 2 to 6 inches, 6 to 12 inches, 12 to 18 inches, 18 to 24 inches, 24 to 36 inches, 36 to 48 inches, and 48 to 60 inches. Approximately 4 grams from each increment was oven dried and scanned for Ru^{106} using a single-channel gamma analyzer.

Fig. 3. Map of White Oak Lake Bed, Showing Transverse Lines of Boreholes
Along Tracts of Ruthenium Seepage

Results

Results from counting the samples were plotted on cross sections to indicate the vertical and lateral distribution of activity. For cross section B-B' (See Fig. 4.) the highest concentrations of activity occur in the uppermost few inches of soil near boreholes 2 and 3. This condition is as expected, since the surface flow of waste at this cross section is largely confined to this zone. Although there was some subsurface movement of waste toward borehole 1, the contamination has spread more in the direction of boreholes 4 and 5, or toward White Oak Creek. This is believed to be due largely to the relatively steep ground-water gradient toward White Oak Creek in the vicinity of this cross section which causes waste to move more readily through the soil.

At cross section D-D' (See Fig. 5.) the maximum concentrations of ruthenium are around boreholes 2, 3, and 5. Surface water flow, which is divided into two prongs at this point, coincides with these zones of maximum concentration. In general, the ruthenium does not enter the lake bed soil over the full length of the section but is limited to these two zones of seepage. The activity observed in boreholes 1, 6, and 7 illustrates the extent of vertical and lateral migration of the ruthenium from the points of initial entry into the soil. In comparing cross sections B-B' and D-D' it is evident that the zone of contamination is wider at the latter than at the former, and that the maximum concentrations are greater at cross section B-B', which is about 400 feet nearer to the source of contamination. The deeper penetration of ruthenium activity at section D-D' is due probably to a slightly greater permeability of the soil at that point.

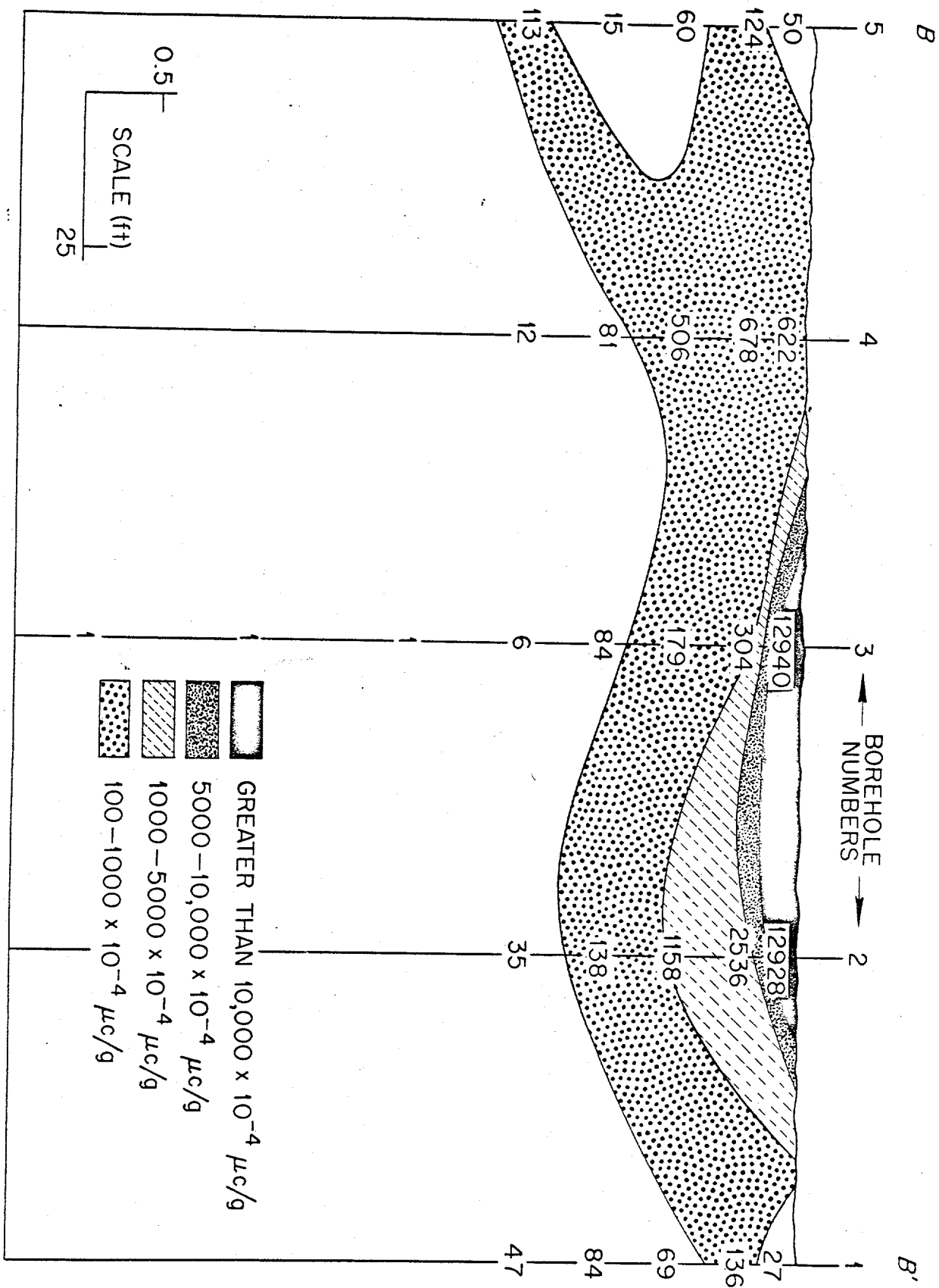


Fig. 4. Cross Section B-B' in White Oak Lake Bed, Showing Ru¹⁰⁶ Concentrations

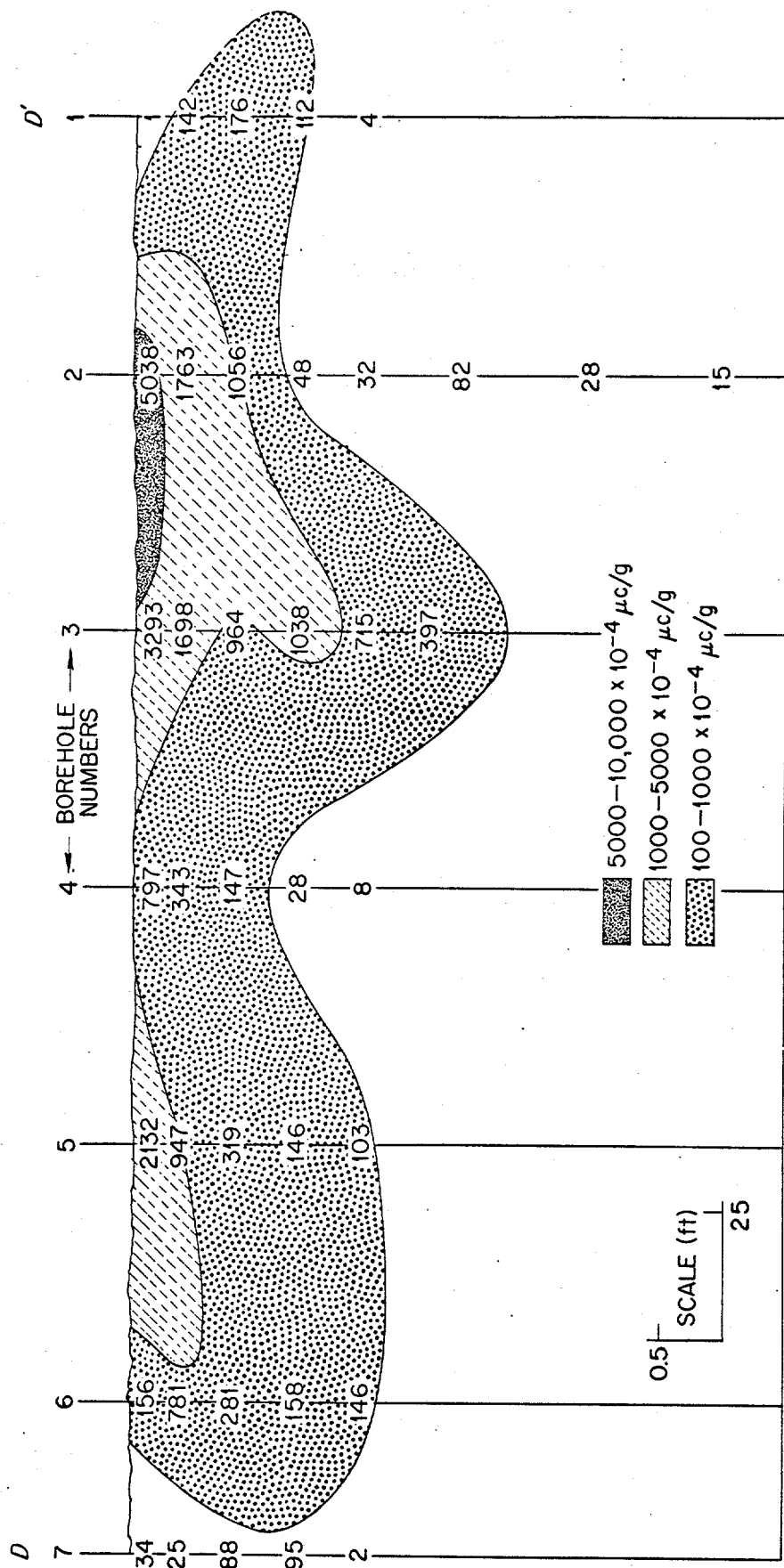


Fig. 5. Cross Section D-D' in White Oak Lake Bed, Showing Ru¹⁰⁶ Concentrations

The same general pattern of Ru^{106} distribution exists at cross section J-J' along Tract 2 (See Fig. 6.) as for cross sections B-B' and D-D' along Tract 1. However, the contamination zone is not nearly as extensive.

The quantity of ruthenium in each cross section was calculated and the results, expressed in curies, summed to yield the total ruthenium in each tract. As of February 1962, Tracts 1 and 2 contained approximately 1200 curies of ruthenium. The quantity of ruthenium for the several increments of depth and the activity detected between cross sections is summarized in Table 1. It is of interest to note that 85% of the ruthenium was found along Tract 1. This is due in part to the larger percentage of ruthenium transport onto the lake bed from the east stream (85% of the total in 1960 and 60% in 1961).

It is probable that the amount of ruthenium in the lake bed varies greatly during the year. The amount of activity leaving the waste pits is largely dependent on the concentration and total volume of ruthenium pumped to the system.⁶ Thus, large changes in the volume and concentration of waste transported to the waste pits will greatly affect the amount of ruthenium that flows onto the lake bed. In addition, less ruthenium is removed from the waste during the wet winter months when most of the water flows over the surface of the bed into the creek than during the dry summer months when the waste streams recharge the ground water in the area. Finally, the short half life of the isotope (1 year) will cause the amount of ruthenium in storage and in transit through the area to change greatly in relatively short periods of time.

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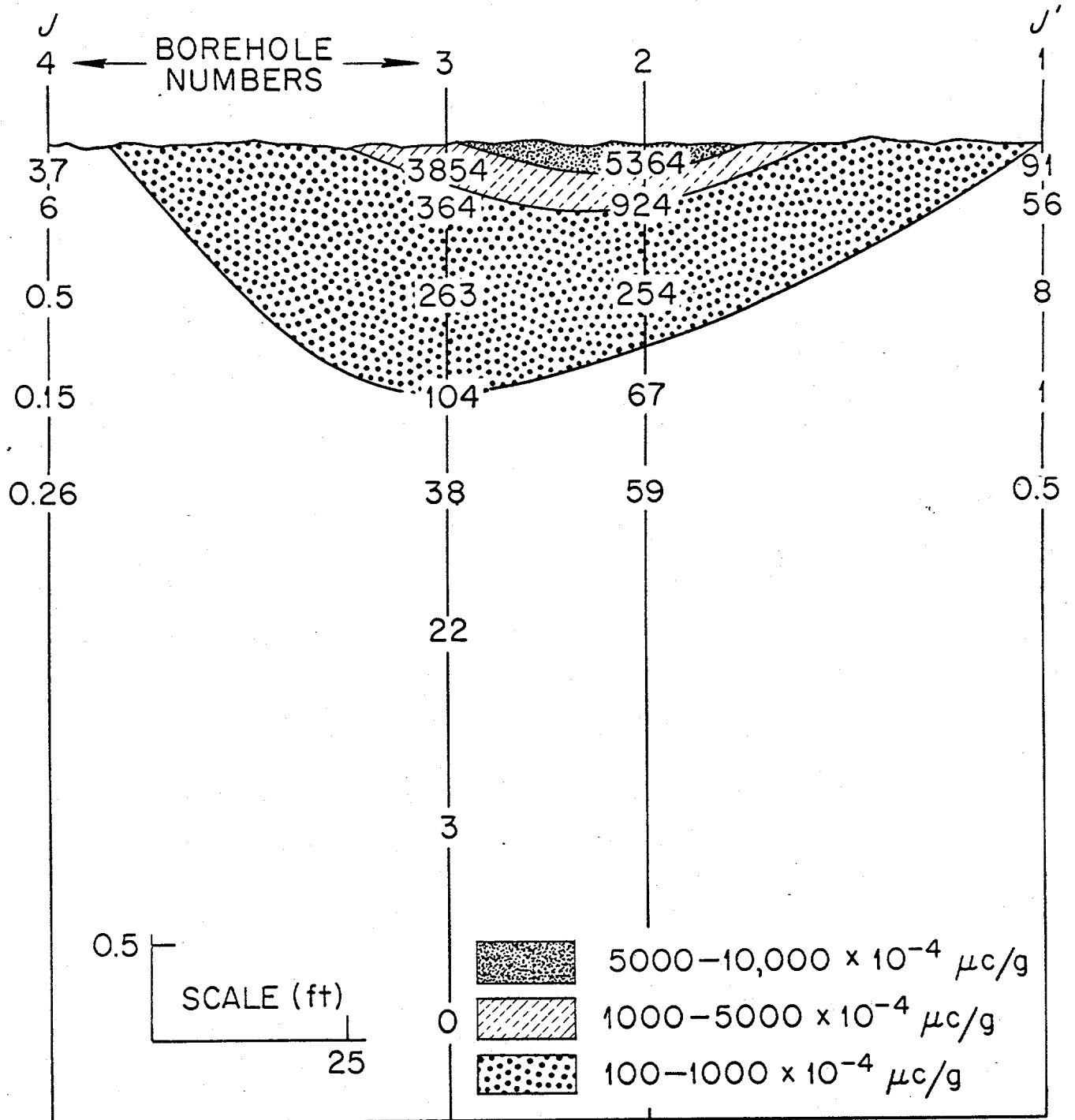


Fig. 6. Cross Section J-J' in White Oak Lake Bed, Showing Ru^{106} Concentrations

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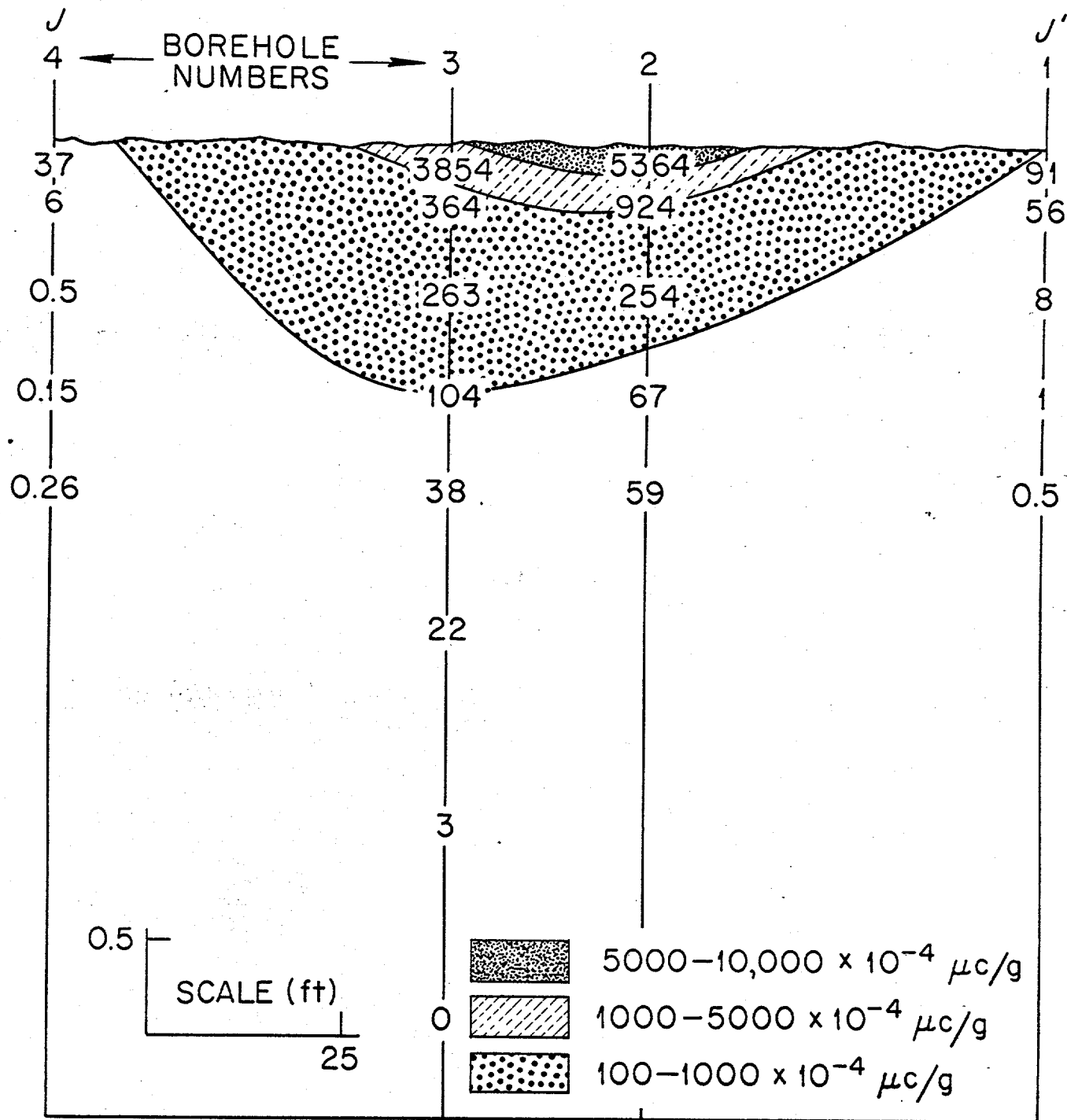


Fig. 6. Cross Section J-J' in White Oak Lake Bed, Showing Ru^{106} Concentrations

Table 1. Distribution of Ru-106 (in curies) in the bed of former White Oak Lake

Cross Sections	Depth from Surface (Inches)						Total
	0-2"	2-6"	6-12"	12-18"	18-24"	24-60"	
Tract 1							
AA'-BB'	62	35	17	3	1	5	123
BB'-CC'	54	32	28	6	4	43	167
CC'-DD'	35	47	39	20	15	99	255
DD'-EE'	27	38	27	19	14	82	207
EE'-FF'	17	28	24	14	6	54	143
FF'-GG'	<u>21</u>	<u>29</u>	<u>27</u>	<u>17</u>	<u>10</u>	<u>37</u>	<u>141</u>
	216	209	162	79	50	320	1036
Tract 2							
HH'-JJ'	5	2	2	< 1	< 1	3	12
JJ'-KK'	13	8	7	2	1	5	36
KK'-LL'	20	19	16	6	1	3	65
LL'-MM'	<u>14</u>	<u>16</u>	<u>12</u>	<u>5</u>	<u>1</u>	<u>11</u>	<u>59</u>
	52	45	37	13	3	22	172
Totals	268	254	199	92	53	342	1208

Retention of Ruthenium by Lake Bed Soil

In order to determine the degree of fixation of ruthenium by lake bed soil, surface samples (0 to 3 in.) of contaminated soil were leached with various solutions in the laboratory. Five-gram portions of oven-dried soil were mixed with 30-milliliter aliquots of various concentrations of KMnO_4 , NaOH , HCl , HNO_3 , as well as with tap water and distilled water. A summary of the per cent of ruthenium leached from the soil by the various solutions is presented in Table 2. Relatively small amounts of ruthenium were leached by tap water, distilled water, and low normality acidic and basic solutions. The quantity of activity removed from the soil increased markedly only when the pH of the leaching solution was raised or lowered beyond the limits of percolating ground water or surface water. Thus, only a small fraction of the ruthenium in the upper few inches of lake bed soil would be expected to move through the soil by downward percolating rainfall or carried off in solution by surface water, which on occasion floods the area.

The mechanisms by which ruthenium is retained by the soil and the identification of the various chemical species of ruthenium that would be expected in the lake bed have been discussed by Gaillardreau.⁷ His work suggests that the ruthenium seeping from the waste pits is a mixture of nitrosylruthenium hydroxide and mononitrato nitrosylruthenium. The hydroxide form is believed to be retained by the soil, while the mononitrato nitrosylruthenium remains essentially unadsorbed. However, as the waste solution travels through the soil toward White Oak Creek, a portion of the mononitrato ruthenium may be converted slowly to the hydroxide to re-establish the equilibrium disturbed by the removal of the hydroxyl complex.

Table 2. Leaching of ruthenium from lake bed soil*

Normality	Solution											
	HCl			HNO ₃			NaOH			KMNO ₄		
	Equilibrium pH	% Leached	Equilibrium pH	Equilibrium pH	% Leached	Equilibrium pH	Equilibrium pH	% Leached	Equilibrium pH	Equilibrium % Leached	Equilibrium pH	% Leached
10 ⁻³	8.20	16	7.90	7.90	15	8.65	8.65	25	7.75	23	7.75	23
10 ⁻²	7.55	14	7.82	7.82	13	11.65	11.65	41	7.95	28	7.95	28
10 ⁻¹	7.00	15	6.53	6.53	14	12.25	12.25	55	9.17	35	9.17	35
1	.50	48	.38	.38	44	12.15	12.15	59	8.80	45	8.80	45
2	< 1	53	< 1	< 1	54	11.90	11.90	60				
3	< 1	58	< 1	< 1	59	11.80	11.80	60				
4	< 1	62	< 1	< 1	61	11.70	11.70	61				
5	< 1	66	< 1	< 1	63	11.25	11.25	61				
6	< 1	69	< 1	< 1	66	11.35	11.35	61				

Tap water 9% leached

Distilled water 9% leached

* Five-gram samples of oven-dried soil leached with 30 ml of solution indicated.

Conclusions

Ruthenium is transported onto the bed of former White Oak Lake by two surface streams that drain the intermediate-level waste pit area. During the dry summer months these streams recharge the ground water in the lake bed, and there is relatively little surface flow into White Oak Creek. However, in the wet winter season much of the waste water from the pits flows over the lake bed into the creek. Thus, in the dry summer season only a small part of the ruthenium that leaves the waste pits reaches White Oak Creek, while in the wet winter months a relatively large fraction of it enters the creek.

Upon entering the lake bed soil, most of the ruthenium is sorbed on the upper few inches of soil. The unsorbed ruthenium moves along the paths predicted by ground-water contours. The configuration of the water table in the area is measurably affected by the flow of the two surface streams that drain the waste pits, causing the ruthenium to migrate underground in many directions, although always toward points of lower elevation. The maximum rate of ground-water movement, and thus of ruthenium, in the upper 2 feet of soil, is believed to be about 5 feet per day; while in the material 2 to 5 feet below the surface, the maximum rate of travel is approximately 0.25 ft per day. Since water moves much more readily through the upper 2 feet of lake bed soil, it is to be expected that a large part of the ruthenium would be restricted in this zone. This was confirmed by analytical data which shows that approximately 70% of the ruthenium in the lake bed is associated with the uppermost 2 feet of soil.

The ruthenium sorbed by lake bed soil moves at such a slow rate through the soil and subsurface travel distance is so great that radioactive decay

reduces the concentration of ruthenium in water reaching the creek to insignificant proportions.

As of February 1962, the lake bed contained approximately 1200 curies of ruthenium. However, it is likely that the amount of ruthenium in the lake bed will vary greatly during the year.

Most of the ruthenium sorbed on the upper few inches of lake bed soil is not readily leachable, and it is unlikely that large amounts of ruthenium would move suddenly through the soil by downward percolating rainfall or be "carried off" in solution by the surface water which occasionally floods the area.

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